Lamont Geological Observatory

Columbia University

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Low Level Alpha Counting of Solids by the Scintillation Method

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by

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Heinrich D. Holland and J. Laurence Kulp

Introduction

Many types of radiation cause emission of light when they impinge on suitable substances. In most instances the flash thus produced has a duration of 10^{-7} to 10^{-5} seconds and an intensity which is a function of the energy absorbed by the fluorescing substance.[1] The development of modern photomultiplier tubes permits the detection of such flashes of light. In the photomultiplier tube the light flash strikes a photo-sensitive surface from which electrons are emitted, forming a pulse which can be amplified electronically and recorded on a mechanical or tape register. Such a radiation detector is known as a scintillation counter.

Since 1940 considerable advances have been made in the design of scintillation counters. Several types of photomultiplier tubes which are superior to older models in amplification, signal to noise ratio, and photosensitive area have been developed.[2][3] Extensive improvements of the phosphors have also made possible the detection of low energy radiation by means of scintillation counting.[4-7]* Modification of the electronic circuitry has increased the field of applicability to include coincidence and anticoincidence work[8-10], beta and gamma ray spectrometry[11] and geologic prospecting[12]. Scintillation counting has thus taken its place in the field of radiation

* for a more complete list of references see Goldsmith, H.H., "Biography on radiation detection", Nucleonics, 4, 142-150, 1949

detection as a useful tool with a wide range of applicability.

The quantitative measurement of the alpha activity of geologic materials has many uses. Most sediments, rocks, and minerals are of low activity and require equipment of high efficiency, low background, and good stability for the accurate determination of their alpha activity. It appeared that in this application scintillation counting would have considerable advantages over the older parallel plate ionization chamber methods. The scintillation counter is not subject to microphonics, has very low background, and can be operated at reasonably high efficiencies. Furthermore no waiting time is required for the adjustment to equilibrium of a filling gas.

This paper describes a type of scintillation counter which has proved desirable for the measurement of the alpha activity of solids. Details of the construction and calibration are given, and the results are compared with that predicted from thick and thin source theory. Later papers will present data on the alpha activity of geologic materials obtained by this type of instrument.

Description of Apparatus

1. General Description

One of the scintillation counters is shown in Plate 1. From left to right the unit consists of the traffic counter register, the housing, and the stack of electronic components. The top level contains the pre-amplifier, timer and regulated high voltage supply. The middle unit is a linear amplifier resting on the scaler. The housing, shown in detail in Figure 1, contains the sample tray, the lucite collimating cone with the phosphor coated on the bottom, and the photomultiplier assembly. Alpha particles emitted upward by the sample strike the phosphor screen. Some of the photons produced by the interaction of the radiation with the phosphor enter the collimating cone and appear at the photocathode of the photomultiplier tube. The photons there eject electrons. The pulse thus formed at the cathode is amplified first by a system of dynodes within the photomultiplier tube and then in the preamplifier and amplifier. In order to reduce the count rate of more active samples sufficiently to avoid overloading of the recorder, the pulses are fed from the amplifier first to the scaler. The output of the scaler then registers on the recorder.

2. The Housing

The housing of a low level scintillation counter should be constructed in such a manner that maximum number of alphas from the sample and minimum number of extraneous impulses reach the photomultiplier tube. The housing shown in Figure 1 fulfills these requirements. The lucite cone, collimates the light impulses from the phosphor coated on the bottom of the cone. Thus alpha pulses from an area of sample much greater than that of the

photomultiplier tube, may be detected. A closely machined depression in the slide, holds the sample tray. Light admission from below is thus prevented during the counting period; an extension of the slide passes under the cone when the sample is removed and hence prevents excitation of the photomultiplier tube while the sample is being changed. Light is prevented from entering the top of the chamber by light-tight seals at the cap of the housing and by the clamping ring, which holds the photomultiplier tube in position. The observation window is desirable to permit positioning of the contact spring.

The housing can be disassembled by unscrewing the three sections of the unit as indicated in the figure. This facilitates changing of the phosphor, cleaning of the housing, and repair work on the cone and photomultiplier assembly.

3. The cone

In order to increase the effective sensitive area of the photomultiplier tube a solid cone of lucite is used. The cone is provided with highly reflective metal coated walls and has a top machined to fit the base of the photomultiplier tube. Photons produced in the phosphor by interaction with an alpha particle can be considered as entering the cone at a point. The fraction of the photons which passes from the cone into the photomultiplier tube is a function of the half-angle of the cone, of the ratio of the area of the base to the top of the cone, and of the solid angle which the photomultiplier tube subtends at the point of entrance of the photons. This latter angle is somewhat larger for photons entering the cone at the center of the base

than for those entering near the outer surface. Thus an alpha particle impinging on the phosphor at the center of the cone will produce an electronic pulse in the photomultiplier tube greater than that produced by an alpha particle of equal energy impinging on the phosphor near the edge of the cone. This limits the feasible base area of the cone, since, when the solid angle subtended by the photomultiplier tube for photons entering the cone at the edge is very small, the pulse produced in the photomultiplier can not be differentiated from the background noise pulses generated in the tube. The most efficient cone in use has a height of 4" and a base diameter of 5". A larger cone 4" high and with a base diameter of 8" was constructed and yielded more net total counts, as would be expected from the above argument. It was found however, that at the normal operating point the total number of counts per cm^2 is less for the larger than for the smaller cone, and that the background increases somewhat with the area of the cone base.

4. The Phosphor

In the present work two commercially produced silver activated zinc sulfide phosphors, the Patterson Type D, manufactured by a DuPont subsidiary, and R.C.A.#33-Z-20A phosphor have been used. Both have proved satisfactory, qualitative tests indicating a slight superiority of the R.C.A. product. AnS-Ag phosphor has been found to have an efficiency of 28% in converting the energy of alpha particles into light energy[5]. This exceeds the efficiency of other commercially produced phosphors for alpha particle particle detection[7]. The light emitted by ZnS-Ag phosphor has an intensity maximum at a

wavelength of 4500 Å, not far from the peak response wavelength of 4750 Å of the R.C.A.#5819 photomultiplier tube. The phosphor is readily available in the form of small crystals which show relatively little self-adsorption of green light.

The only direct disadvantage of the ZnS-Ag phosphor is the long-term phosphorescence of the material. Figure 2 shows the count rate as a function of time after a long exposure of the phosphor to light. This data indicates that the majority of the phosphorescence effect disappears in three hours and that after eight hours the statistical fluctuations in the count rate obscure the trend of the decay. For the first two hours the count rate is nearly inversely proportional to the time since exposure to light.

Deactivation of the phosphor after exposure to alpha radiation takes place at approximately the same rate as deactivation after exposure to light. However, it was found that after counting freshly ground zircons, the decay of the background counting rate decreases approximately exponentially reaching one half of its original value in about 16 hours as shown in Figure 3. The explanation of this effect is possibly to be found in the liberation of a considerable amount of radon from the freshly broken zircons. The half-life of radon is 3.825 days. However, since diffusion of radon probably took place from the active area of the counter during the time of observation, a shorter apparent half-life is to be expected in the present case.

The phosphor is applied to the base of the cone by shaking an

excess of phosphor crystals on the surface freshly coated with a thin layer of stopcock grease. The quantity of phosphor readily taken up by the grease depends on the thickness and nature of the grease. The nature of the phosphor screen affects the efficiency of the counter to an appreciable extent. Hence each time a new coat of phosphor is applied to the cone, the counter must be calibrated. A transparent coat of grease just thick enough to ensure complete coverage of the base of the cone by one layer of phosphor crystals appears to yield the highest efficiency.

5. The Photomultiplier Tube

For low activity counting in conjunction with a cone an end window tube with a large photosensitive area is most suitable. These features are combined in the RCA #5819 photomultiplier. The #4588 and #5311 photomultiplier tubes manufactured by Electrical and Musical Industries, Great Britain, should also prove useful for this work[(2)].

6. The Electronic Components

As indicated in Plate 1 an amplifier, a scaler, a register, and a high voltage supply are the necessary components of a low level scintillation counter. Several combinations of commercial units have been tried and found satisfactory for this purpose. In one of these counting units the signal passes from the photomultiplier tube into a Model 205 preamplifier, thence into a

Model 204 C amplifier and a Model 110 decade scaler, all products of the Atomic Instrument Co., Boston, Mass.

The pulse from the scaler is fed into a Model-SCI-2 Streeter-Amet Traficounter which automatically prints the accumulated count at 15 minute intervals. A Model 305 power supply of the Atomic Instrument Co. furnishes the high voltage for the photomultiplier tube assembly. The voltage for the individual dynodes of the photomultiplier tube is obtained by means of the voltage divider shown in Figure 4.

The above unit has consistently given good service. However, the expense of the unit is considerable, and fluctuations of up to 10% in the count rate due to the instruments have been noticed during a 24 hour counting period. The fluctuations are probably due mainly to voltage drifts and to drifts in the discrimination circuit. These defects may be removed in the forthcoming Atomic Instrument Model 1040X scaler which combines the preamplifier, amplifier, scaler, and high voltage supply in one shassis, and which will provide more adequate control of the photomultiplier tube voltage.

In another counting arrangement the place of the "Atomic" Instruments is taken by a Nuclear Instrument and Chemical Corp. Model 162 scaler. However the discriminator level of this unit is not precision calibrated and the stability of the high voltage supply does not appear to be sufficient to obtain high precision

data in this type of counting.

The electronic components in the third counter consist of a Model 206 preamplifier and Model 204 B amplifier of the Atomic Instrument Co. in conjunction with a Nucleonic Corp. scaler. The high voltage control of the latter unit has shown the greatest variations of the units which have been placed in service.

The Traficounters have been invaluable in detecting spurious counts and long-term fluctuations in the counting rate. Cyclotron Specialties registers have been in use for short-term experiments with samples of high activity.

III. Thin and Thick Source Counting with the Scintillation Counter

In order to determine the absolute alpha activity of a sample, it is necessary to correct the measured count rate. Factors which affect the observed count are: the nature of the sample, the distance between the sample and the phosphor screen, the photomultiplier sensitivity, and the circuit variables. In the paragraphs to follow equations relating the observed count rate to the absolute alpha activity are developed. The experimental data are then analyzed and shown to be consistant with the theoretically derived relationships.

1. General Theory of Thin and Thick Source Counting

Since the range of alpha particles in solids is very small, the distance between the nucleus emitting the alpha particle and the phosphor screen determines to a large extent the energy of the alpha particles which reach the phosphor. If the energy of an alpha particle omitted at the base of a source is not appreciable decreased upon traversing the sample, the source is defined as "thin". If the alpha particles emitted at the base of the sample are stopped completely before reaching the surface of the sample, the source is considered "thick".

Consider a unit volume of area dA and thickness dy at a depth y in a radioactive source. The number of alpha particles

from this unit volume which are counted per unit time is a function of

a) N, the number of alpha particles per cubic centimeter which are emitted per hour by the source.

b) E, the energy in M.E.V. of the alpha particles emitted by the unit volume; the energy is related to R, the air-range in cm of the alpha particles. A commonly used approximation is the equation

$$R = bE^{3/2}$$

where b is a proportionality constant.

c) the stopping power of the source material, which can be expressed in terms of m, the ratio of the range in the source material to the range in air of alpha particles emitted within the source.

d) a, the distance in cm from the source to the phosphor screen.

From the unit volume N.dA.dy alpha particles are emitted per hour. Only those alpha particles are detected by the counting system which reach the phosphor with a residual range greater or equal to r cm in air. From the relations in Figure 5 it can be seen that alpha particles emitted vertically upward have a residual range of $(R - a - y/m)$ air-cm. upon reaching the phosphor; alpha particles emitted at an angle θ from the vertical will have a residual range of $[R - (a+y/m)/\cos\theta]$ air-cm. It follows that, excepting edge effects, those alpha will reach the phosphor with sufficient energy to be recorded, which are emitted from the unit volume at an angle θ from the vertical such that

$$\theta \leq \cos^{-1} \frac{(a + y/m)}{R - r} \qquad (1)$$

Since the direction of emission of alpha particles from the unit volume is randomly distributed in space, the number of alpha particles passing per unit time through equal areas of a sphere surrounding the unit volume will be equal. It can be shown (13) that the fraction of the area of a sphere which is intercepted by a cone of half-angle θ is

$$\frac{1}{2}(1 - \cos\theta)$$

Thus the fraction, F, of the total number of alpha particles emitted from the unit volume which is counted will be

$$F = \frac{1}{2}\left(1 - \frac{a + y/m}{R - r}\right) \tag{2}$$

The actual number of alpha particles, dn which are c ounted per hour from the unit volume will be F.N.dA.dy; thus

$$dn = \frac{N}{2}\left(1 - \frac{a + y/m}{R - r}\right) dA \cdot dy \tag{3}$$

If the source is "thick", the number of alpha particles, n, which are counted per hour from the whole source will be

$$n = \int dn = \frac{NA}{2}\int_0^{m(R-r-a)} \left(1 - \frac{a + y/m}{R - r}\right) dy \tag{4}$$

or

$$n = \frac{NAm}{4}\frac{(R-r-a)^2}{(R-r)} \tag{4a}$$

If the thickness, t, of the source is less than m(R-r-a) cm, i.e. if the source is thin enough so that at least a part of the alpha particles emitted at the base of the source are recorded,

then t replaces m(R-r-a) as the upper limit of integration in equation (4). Consequently the count rate, n, will follow the relation

$$n = \frac{NAt\left[2m(R-r-a)-t\right]}{4m(R-r)} \qquad (5)$$

In such a source the count rate recorded is a function of the thickness t. When t is very much smaller than m(R-r-a) cm, i.e., when the absorption in the source of the energy of the alpha particles is negligible equation, (5) reduces to the form

$$n = \frac{NAt\ (R-r-a)}{2\ (R-r)} \qquad (6)$$

If furthermore the air-gap between the sample and the phosphor is small, so that "a" is much smaller than (R-r), equation (6) further reduces to the form

$$n = \frac{NAt}{2} \qquad (7)$$

In this case one half of all the alpha particles emitted by the source are registered.

In the above calculations it was assumed that the phosphor screen is infinitely extended, so that all alpha particles, regardless of their point of origin, are counted provided their residual range on reaching the phosphor is greater than r air-cm. In practice the area of the phosphor screen has been made equal to that of the sample. Some alpha particles emitted near the edge of the sample tray at angles close to the horizontal therefore fail to impinge on the phosphor. It can be shown that for thick sources of the size used in the present experiments the effect of this loss is less than

1% of the total count rate provided the air distance between sample and phosphor is less than 0.2 cm.

2. Experimental Results

(i) Thin sources

Thin sources were prepared by plating on the order of 10^{-12} gm of polonium on grade A nickel plates 1 inch square and 0.04" thick from a slightly acid solution of polonium chloride.

One of these sources was placed at the center of the sample tray at a distance of about 0.2 cm from the phosphor screen, and the count rate was determined as a function of voltage and discriminator level setting. The data thus obtained are summarized in Figure 6. From right to left at high voltages the curves can be divided into three regions:

a) a sharp rise of the count rate at high discriminator level settings,

b) a flat region at intermediate discriminator level settings, and

c) a sharp rise of the count rate at small discriminator level settings.

It can be seen that, as the voltage across the photomultiplier assembly is increased, the flat central portion of the curve becomes more pronounced, the curve becomes steeper at high discriminator level settings, and the inflection point in the lower range of discriminator level settings moves toward the right. Since the polonium source used was extremely thin, alpha particles leaving the source into the air space between the source and the phosphor must have been essentially

monoenergetic except for straggling and backscattering. However, since the source was not in direct contact with the phosphor, alpha particles emitted at the same point in the source but at different angles with the vertical traveled different air-distances before impinging on the phosphor. The residual range on reaching the phosphor was therefore not the same for all of the alphas emitted by the source. This effect on the residual range distribution for polonium alpha particles impinging on the phosphor was calculated for a = 0.2 air-cm and is shown in Figure 7. From this Figure it can be seen that 97% of the alpha particles emitted by the source reach the phosphor and that 80% of the alphas arrive with more than 86% of their original energy. Since the light energy liberated in the phosphor is proportional to the residual energy of the alpha particles impinging on the phosphor, the distribution of the intensity of the light flashes should follow approximately the distribution of the residual range of Figure 6, provided that a sufficient thickness of phosphor is available to absorb all the residual energy of the alpha particles. The difference in the percentage of light which enters the photomultiplier tube from flashes of identical intensity at different parts of the base of the cone will cause the pulse height distribution curve in the photomultiplier tube to have a slightly less promounced curvature than the theoretical residual range distribution curve of Figure 7.

The discriminator level setting on the scaler indicates the minimum pulse height necessary for registering a signal in the recording system. Thus curves of count rate versus discriminator level setting indicate the pulse height distribution of signals from the

the photomultiplier tube. In terms of these concepts the curves of Figure 6 can be interpreted. The count rate versus discriminator level setting curve at 1200 volts approximates the expected distribution except at very low and at very high discriminator level settings. The sharp rise at low discriminator level settings seems to be due to secondary flashes of light produced in the phosphor after the main burst due to the collision of alpha particles with particles of phosphor. The secondary flashes appear to be very numerous and quite small compared to the primary flash. At very high discriminator level settings the count rate drops more rapidly than is predicted on the basis of the simple theory. It is possible that the steeper slope is due to non-linearity of response in the electronic system at alpha energies above 3 Mev.

At voltages lower than 1200 volts the plateau progressively decreases in length. Since the light distribution reaching the photomultiplier tube is not affected by the voltage applied to the photomultiplier assembly, changes in the curve of count rate versus discriminator level setting must be due to changes in the sensitivity of the photomultiplier tube. It appears that the change in the sensitivity of the tube with voltage is not constant over the entire photosensitive area, for, if the change of sensitivity were constant over the whole area, a decrease in the voltage would cause a uniform contraction of each portion of the curve. That this is not the case is shown especially by the disappearance of the plateau and by the pronounced downward curvature of the right hand part of the curves at low voltages.

If secondary pulses are neglected, the extrapolation of all the curves to zero pulse height yields almost the same value. This indicates that the extrapolated count rate at zero discriminator level setting is the actual rate at which alpha particles are impinging on the phosphor. This number is the same regardless of the voltage applied to the photomultiplier tube.

On the basis of this discussion it appears probable that if less than 1200 volts are applied to the photomultiplier assembly, the photocathode of the photomultiplier tube is not uniformly sensitive. Hence changes in voltage differently affect each section of the photocathode and it is only in the vicinity of 1200 volts that the sensitivity becomes uniform over the whole photosensitive area.

The effect on the count rate of varying the effective distance between the sample and the phosphor was determined by inserting thin sheets of aluminum foil between the sample and the phosphor. Figures 8, 9, and 10 indicate the data obtained under these conditions for a polonium source of somewhat higher activity than that used in the above experiments. As in the previous experiments the expected relationship of count rate to discriminator level setting was obtained only at high voltages. The shape of the curves is seen to be independent of the thickness of aluminum foil interposed between sample and phosphor, and is therefore independent of the energy of the particles impinging on the phosphor. This fact was borne out by experiments on thick sources discussed below and points to the sensitivity of the photomultiplier tube as the prime factor in determining the shape of the count rate versus discriminator level setting curve at voltages lower than 1200 volts.

It is of interest to note that each of the curves of count rate versus discriminator level setting intersects the ordinate at approximately the same point when extrapolated. The relation of the experimentally determined values of the intercept can be checked by means of equation 6.

In the present experiments the distance from the top of the sample to the phosphor was 0.4 cm. If 1.47 mg Al/cm^2 is taken as an average value of the aluminum equivalent for the stopping power of 1 cm of air, the decrease in counting rate due to the presence of the aluminum approximates the expected decrease. A summary of the data is presented in Table I.

Table I

Count rate of thin polonium source as a function of absorber thickness

Absorber thickness mg Al/cm^2	"a" (air-cm)	Count rate experimental		Count rate calculated
0	0.40	24.6 26.3 26.6	25.8	(25.8)
2.45	2.07	11.3 11.4 11.8	11.5	12.5
4.90	3.73	0.48	0.48	0.46

(ii) Sources intermediate between thin and thick sources

According to equation 5 the count rate for sources intermediate

between thin and thick sources in which only one radioisotope is present is

$$n = \frac{NAt\ [2m(R-r-a)-t]}{4m\ (R-r)}$$

The applicability of this equation was tested by recording the count rate of a series of progressively thicker sources of a sample of ocean bottom sediment. The thinnest of these sources was almost a "thin" source, (0.8 mg/cm^2) and the thickest approached a "thick" source (4.0 mg/cm^2) in dimensions.

The alpha particles emitted by the ocean bottom sediment originate in a number of alpha active radioisotopes present in the sample. The range of the alphas emitted by each radioisotope is associated with a distinct range, R_i; thus the count rate, n, is the sum of the contributions from each of the radioisotopes

$$n = \sum_i \frac{N_i\ At\ [2m(R_i - r - a) - t]}{[R_i - r]} \qquad (5a)$$

This can be written in the form

$$n = pt - qt^2 \qquad (5b)$$

where

$$p = A \sum_i \frac{N_i\ (R_i - r - a)}{(R_i - r)}$$

and

$$q = A \sum_i \frac{N_i}{(R_i - r)}$$

The sediment was ground in an agate mortar. A few drops of ethyl alcohol were then added and the mixture was allowed to stand until the coarse material had settled. The supernatant liquid, containing the finer portion of the sample, was decanted into a drying unit consisting of the source plate, a rubber ring and a metal ring $\frac{1}{2}$" high clamped together to prevent seepage of the liquid. The suspension was heated at about 100°C until the alcohol had evaporated. A uniform coating of sample on the brass source plate was usually obtained. The source plate was then inserted into one of the scintillation counters. The voltage across the photomultiplier assembly during the run was approximately 1000 volts.

As can be seen from Figure 11, good agreement between the theoretical curve and the experimental data was obtained. The discrepancies in the data are amply accounted for by inhomogeneous distribution of the sample on the tray, errors in the measurements of the area covered by the sample, non-uniform loss of radom in successive samples, and slight fluctuations in the counting rate due to the instruments.

(iii) Thick sources

The count rate of thick sources was determined under various conditions to test the applicability of thick source theory to counting with the present type of apparatus. The first type of source was uranium metal foil, the second, samples of uraninite. From equation (4a) it is to be expected that the count rate, n,

from each isotope will obey the relation

$$n = \frac{NAm}{4} \frac{(R-r-a)^2}{(R-r)}$$

so that the total count rate, n_t, will be the sum of such terms for U-234, U-235, and U-238. We can therefore write

$$n_t = \sum_{i=1}^{i=3} \frac{N_i A m_i}{4} \frac{(R_i-r-a)^2}{(R_i-r)} \qquad (8)$$

Experiments were conducted with square plates of uranium, 0.18 mm thick, to test the applicability of this equation. It is well-known that the range of alpha particles in a metallic source is proportional to the wquare root of the atomic weight of the source material divided by its density. On the basis of this relation, m, the ratio of the range of alpha particles in uranium to their range in air is 2.30×10^{-4}. The number of disintegrations of U-238 atoms per cubic centimeter can be calculated knowing the decay constant of uranium, and has the value 2.31×10^5 sec^{-1} per cc. If, as is the case in most uranium metal derived from geologically old minerals, U-234 is in equilibrium with U-238, an equal number of alpha particles per second are derived from this isotope. The number of U-234 atoms present in the source is much less than one per cent of the number of U-238 atoms. The U-235 concentration is approximately 0.721%.

On the basis of recent experiments [14] the range in air of U-238 alphas is 2.70 cm, those of U-234, 3.25 cm, and those of U-235, 300 cm.

On the basis of these considerations n_{U_t}, the count rate per min per cm^2 of source is found to be

$$n_{u_t} = n_{u}238 + n_{u}234 + n_{u}235$$

$$= 7.94 \times 10^2 \left[\frac{(2.70-r-a)^2}{(2.70-r)} + \frac{(3.25-r-a)^2}{(3.25-r)} \right]$$

$$+ 0.37 \times 10^2 \frac{(3.00-r-a)^2}{(3.00-r)}$$

Curves of count rate versus discriminator level setting were obtained for sources of uranium at different distances from the phosphor. One of these curves is shown in Figure 12. Extrapolation to zero discriminator level setting yields the count rate at r = 0. Figure 13 shows the value of the count rate calculated on the basis of the above equation and the experimentally determined points. Fairly satisfactory agreement is seen to exist between experiment and theory.

In the uraninite specimens eight alpha emitting elements are present. The alphas from each element are associated with discrete energies so that the total rate, n_t, can be written in the form

$$n_t = \sum_{i=1}^{i=8} \frac{AN_i m}{4} \frac{(R_i - r - a)^2}{(R_i - r)} \qquad (10)$$

In order to test the relationship between the residual alpha energy and the height of the electronic pulse count rate versus discriminator level curves were obtained at 1300 volts across the photomultiplier assembly for a thin polonium source, a thick uranium foil source, and a thick source of uraninite. It was found that the value

of the discriminator level setting at which the count rate had dropped to zero was the same for the three sources. Since the energy of the most energetic alphas from the three sources was not the same, the residual range of these alphas must have differed. It follows that a non-linearity between residual alpha energy and pulse height in the photomultiplier tube exists at alpha energies in excess of 3 Mev under the present operating conditions.

Since the eight alpha emitting elements in old uraninite samples are in radioactive equilibrium, the same number of alpha particles are emitted per unit time from each of the radio-elements. Thus equation (10) can be rewritten in the form

$$n_t = \frac{NAm}{4}\left[\cdot 16a + \sum_{i=1}^{i=8} (R_i - r) + \frac{a^2}{(R_i - r)}\right] \qquad (10a)$$

As in the previous cases "a" represents the total length of path in air-cm between the sample and phosphor. This includes not only the air space between the sample and the phosphor screen but also the air-equivalent of absorbers placed between the sample and the phosphor.

Figures 14 and 15 show the count rate of one of the uraninite specimens as a function of the discriminator level setting with different thicknesses of aluminum foil inserted between the phosphor and the sample. Since the ranges of all the alpha particles emitted by the uraninite are known, the decrease of count rate with absorber thickness at zero absorber thickness can be calculated by means of equation (10) or (10a). Figure 16 shows the calculated curve and also the points derived experimentally from the data of Figures 14

and 15. The experimental points follow the trend of the calculated curve quite well. The difficulty of determining accurately the intercept of the curve of count rate versus discriminator level setting can be held responsible for at least some of the scattering of the experimental points.

Figure 17 shows the variation of count rate with discriminator level setting for a specimen of uraninite at a voltage of 1250 volts across the photomultiplier assembly. As in the case of the thick uranium source there is no plateau in the middle of the range of discriminator level settings. In all other aspects the two curves are also similar as is to be expected from the similar nature of the two sources.

In the above paragraphs a body of data has been presented which is found to be consistent with fundamental concepts. By means of these concepts it is possible to determine the absolute alpha activity of samples.

IV. Determination of Absolute Counting Rates

With the present experimental arrangement close to 100% counting efficiency is reached only when high voltages are applied to the photomultiplier tube. When such voltages are used, the background count rate is much greater than the count rate due to most rocks and minerals. It is therefore necessary to operate at lower voltages, and thus at considerably less than 100% counting efficiencies for these samples. The choice of the actual operating point is determined by the relation of efficiency to background, and by the rate of variation of the count rate with drifts in the circuit variables. It has been found in these experiments that for use with low-activity samples the most favorable tube voltage is between 950 and 1100 volts, depending somewhat on the apparatus and on the photomultiplier tube used. Under these conditions an efficiency of 25% for thick sources at a background counting rate of 30 counts per hour has been maintained consistently, and background counts of 15 counts per hour at 25% efficiency are attainable with due precautions.

In order to determine the absolute count rate of a rock or mineral the relative count rate must be corrected in terms of the counting efficiency of the system at the operating point. A small alpha source of known activity is placed below the cone at several points between the center and the outer edge, and the count rate at the operating point is determined. This procedure is repeated in different directions from the center of the cone.

An average of these values, weighted for the area represented by each position, is computed from this data. In order to relate the efficiency thus determined for the reference source to the efficiency of the system for the samples measured, the constants a and r must be determined. The distance from the reference source to the phosphor can be measured by means of a micrometer. r is then computed by means of equation (4a) if the reference source is a thick source, or by equation (6) if a thin reference source is used. The value of r thus found is applied to calculating the absolute count rate of the samples. In general the values of "a" for the reference source will differ from that for the samples.

Figure 18 shows the counting efficiency for a thick source foil of uranium metal 1.91 cm^2 in area at various positions below the cone shown in Figure 1. Due to the increasingly unfavorable geometry the count rate decreases from the center of the cone outward. There is also considerable difference in the observed count rates at equal distances from the center but in different directions from the center of the cone. These variations are probably due to variations in the thickness of the phosphor, to imperfections in the cone, and to azimuthal variations in the sensitivity of the photomultiplier tube.

Figure 19 shows the effect of position of the source under the cone at 1200 volts on the count rate of a thin source of polonium. The upper curve is a plot of count rate versus discriminator level setting for a small thin polonium source placed beneath the center of the cone. The lower curve shows the count rate when the same sample was placed beneath the edge of the cone.

At high discriminator level settings the curves approach each other. At discriminator level settings between 70 and 90 the curves reach their maximum separation, and at values between 40 and 70 the two curves again approach each other. From this data two effects are noticeable: the loss of alpha particles due to edge effects and the reduction by the geometry at the edge of the cone of light energy reaching the photomultiplier tube. The loss of alpha particles due to edge effects can be ascertained by extrapolating the two curves to zero discriminator level setting, and appears to be between 15% and 20%. The reduction of light energy is witnessed by the reduced slope of the lower curve in the middle region of the discriminator level setting. It is interesting to note that the zero count rate intercept of the two curves does not differ greatly. This may be another indication of the non-linearity of the response of the electronic system at high alpha energies.

A cone larger than the one shown in Figure 1 has been tested in a suitable housing. The height of this cone was 4" and the diameter of the base 8" compared to corresponding measurements of 4" and 5" for the smaller cone. The top diameter was machined to fit the base of the photomultiplier tube. It was found that the efficiency to about 2½" from the center of two cone types were comparable, but that the contribution to the count rate of sample underneath the edge of the large cone is negligible. It thus appears

that extension of the base diameter beyond 5" at a height of 4" and an upper diameter of 2" does not increase the total count rate appreciably.

V. Summary

Scintillation counters have been developed which have advantages over parallel plate ionization chambers for the low level alpha counting of rocks and minerals. The design and construction of the counters in presented. An important feature of the apparatus is a lucite cone which is used to gather light flashes from a much larger surface area of sample than is possible with a photomultiplier tube alone. The activation and deactivation of ZnS-Ag phosphor is discussed, and the electronic components utilized for three scintillation counters are described.

The theory of thin and thick source counting is reviewed, and its application to scintillation counting is developed. The relation between count rate, sample thickness, absorber thickness, and the circuit constants received special attention.

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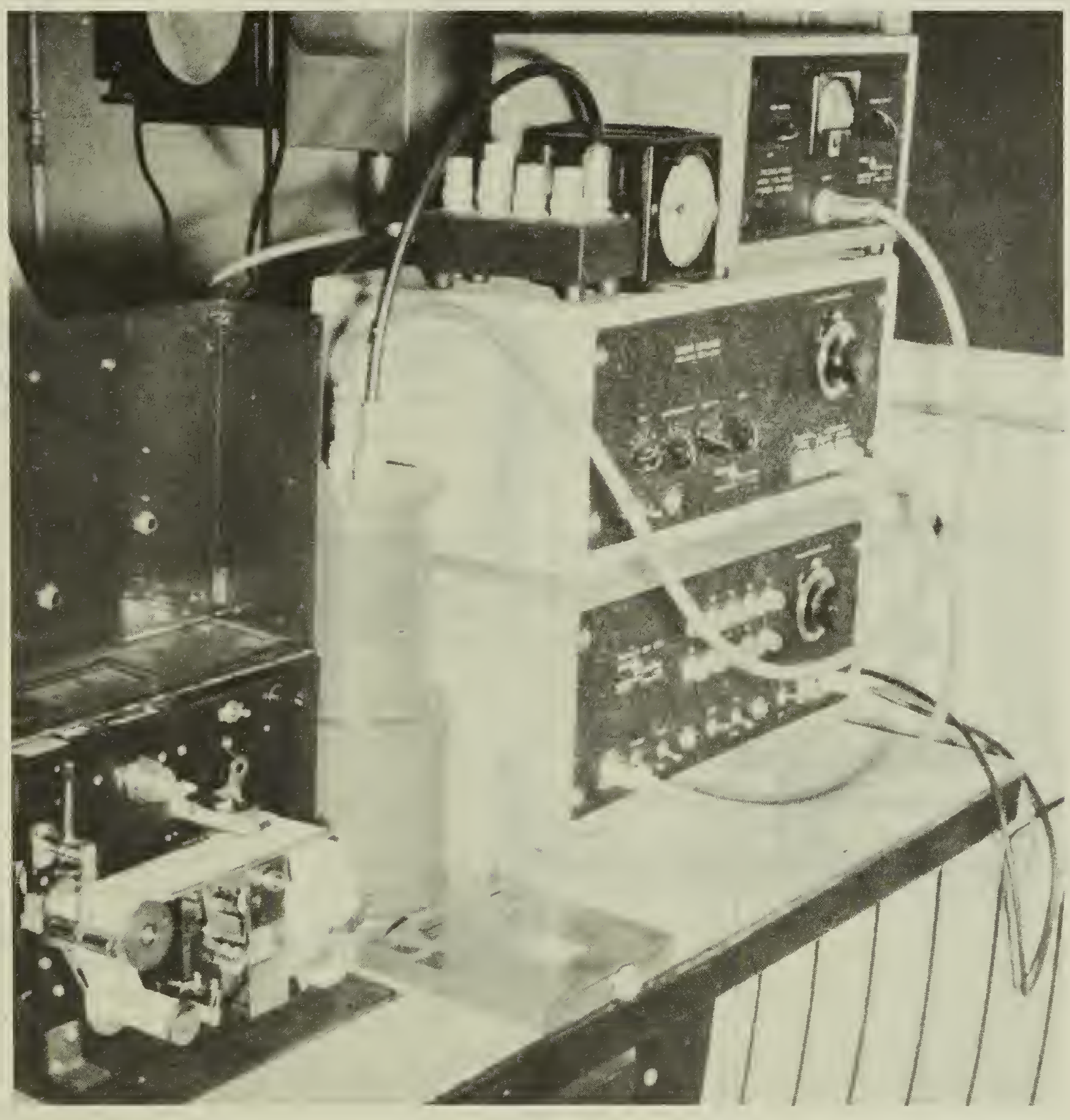

Plate 1. Scintillation Counter Unit.

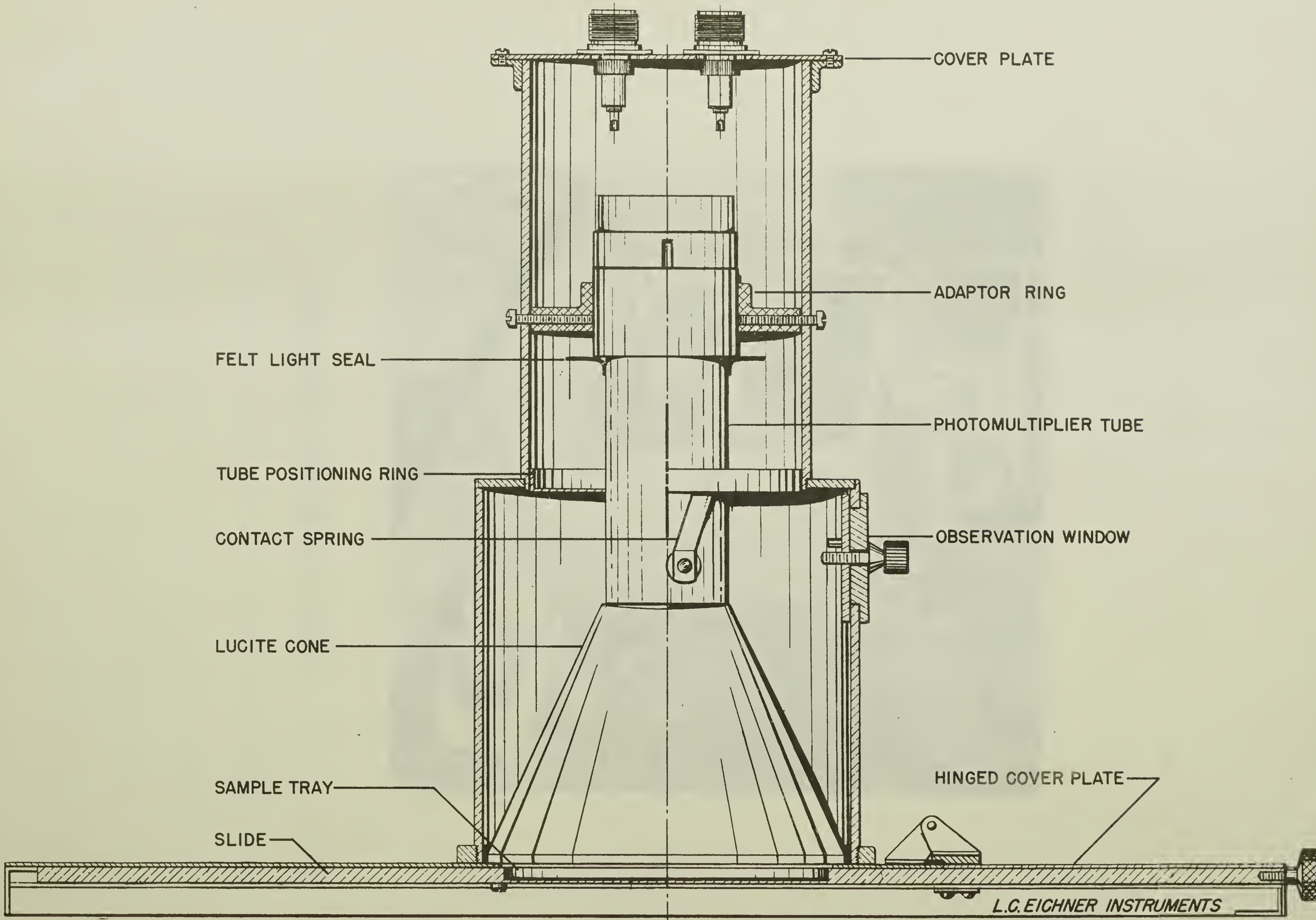

Figure 1. Cross-Section of scintillation counter housing.

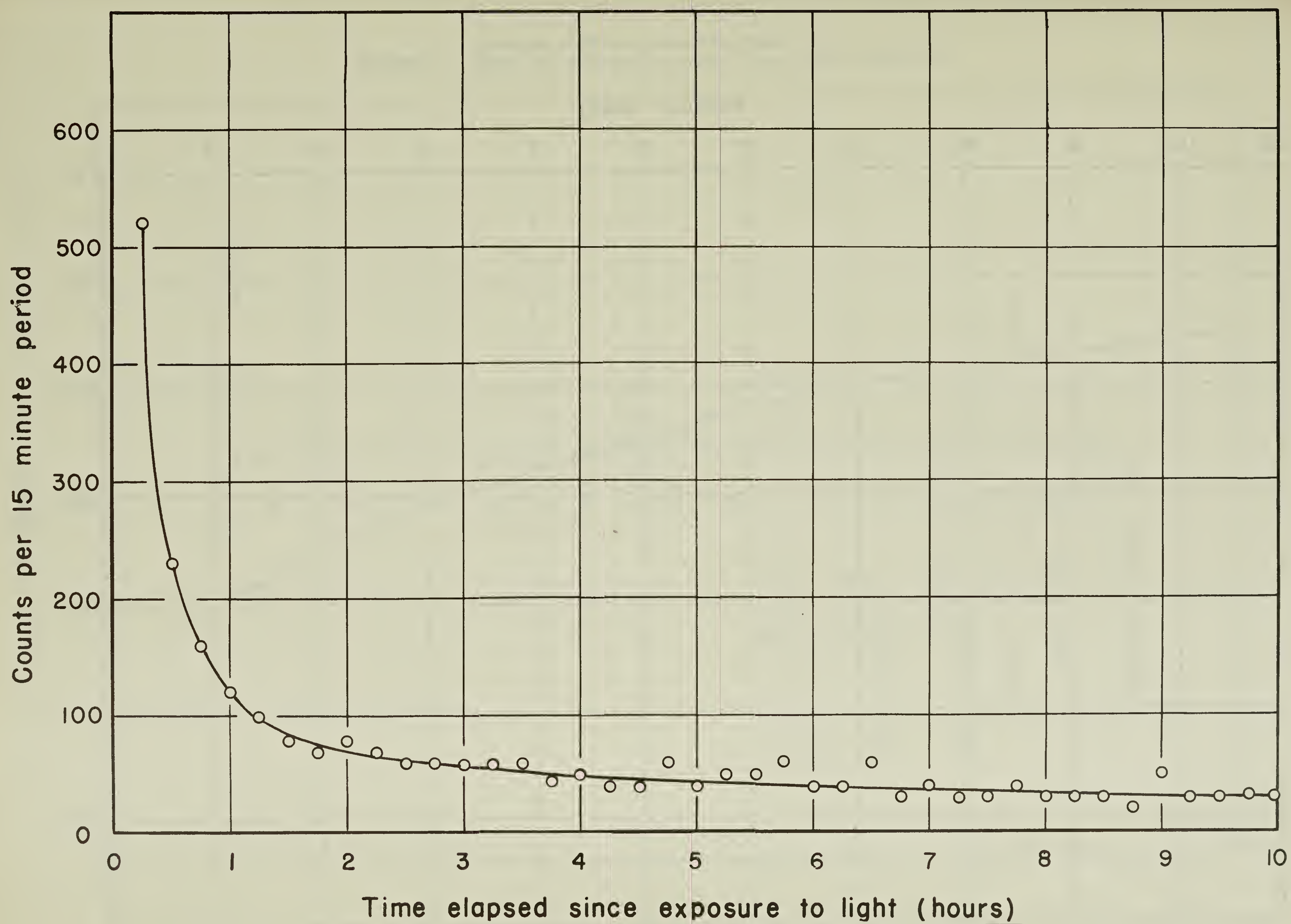

Figure 2. Decay of phosphorescence in ZnS-Ag phosphor after exposure to light.

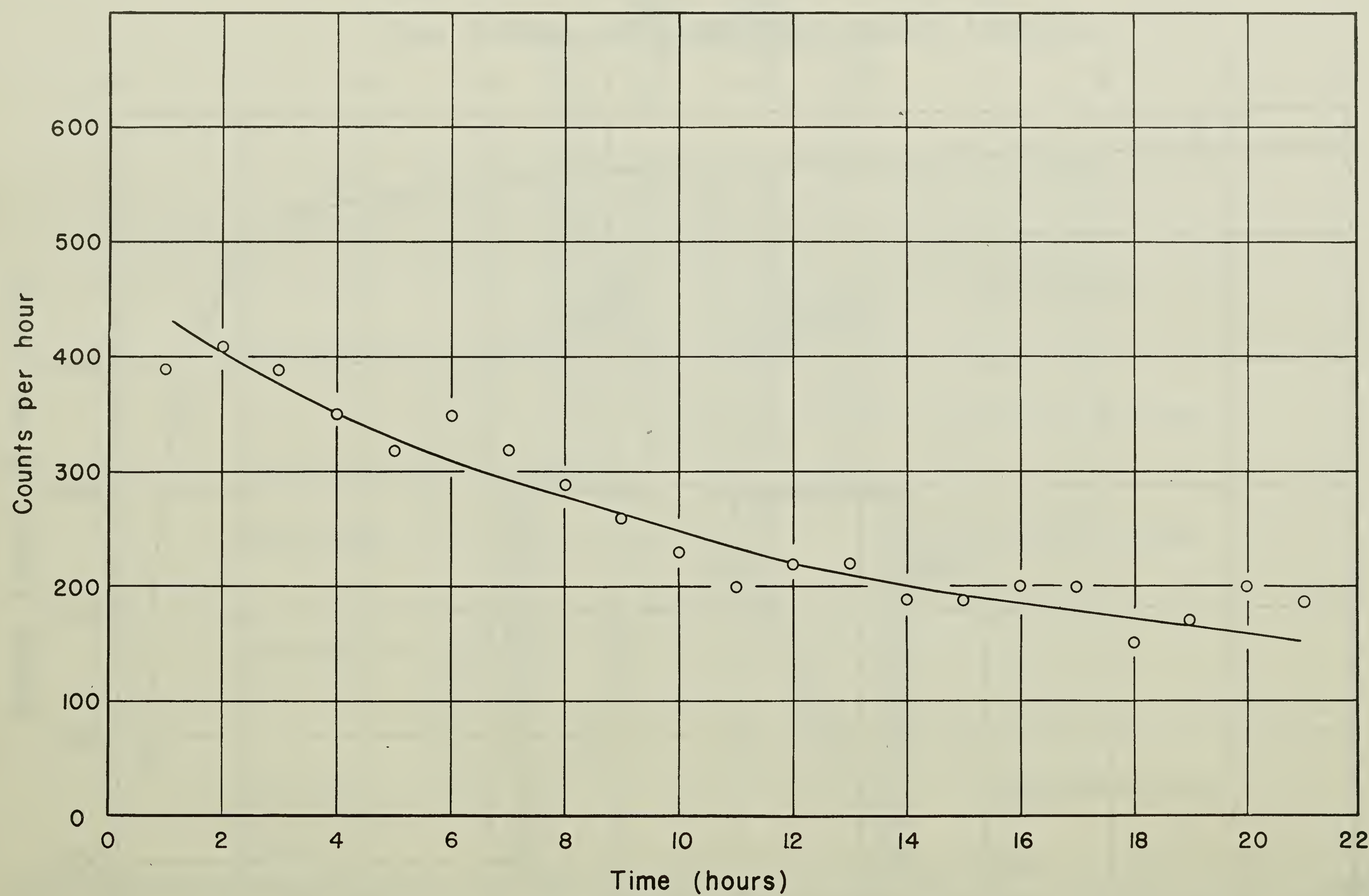

Figure 3. Decay of background count rate after analysing freshly broken zircon specimen.

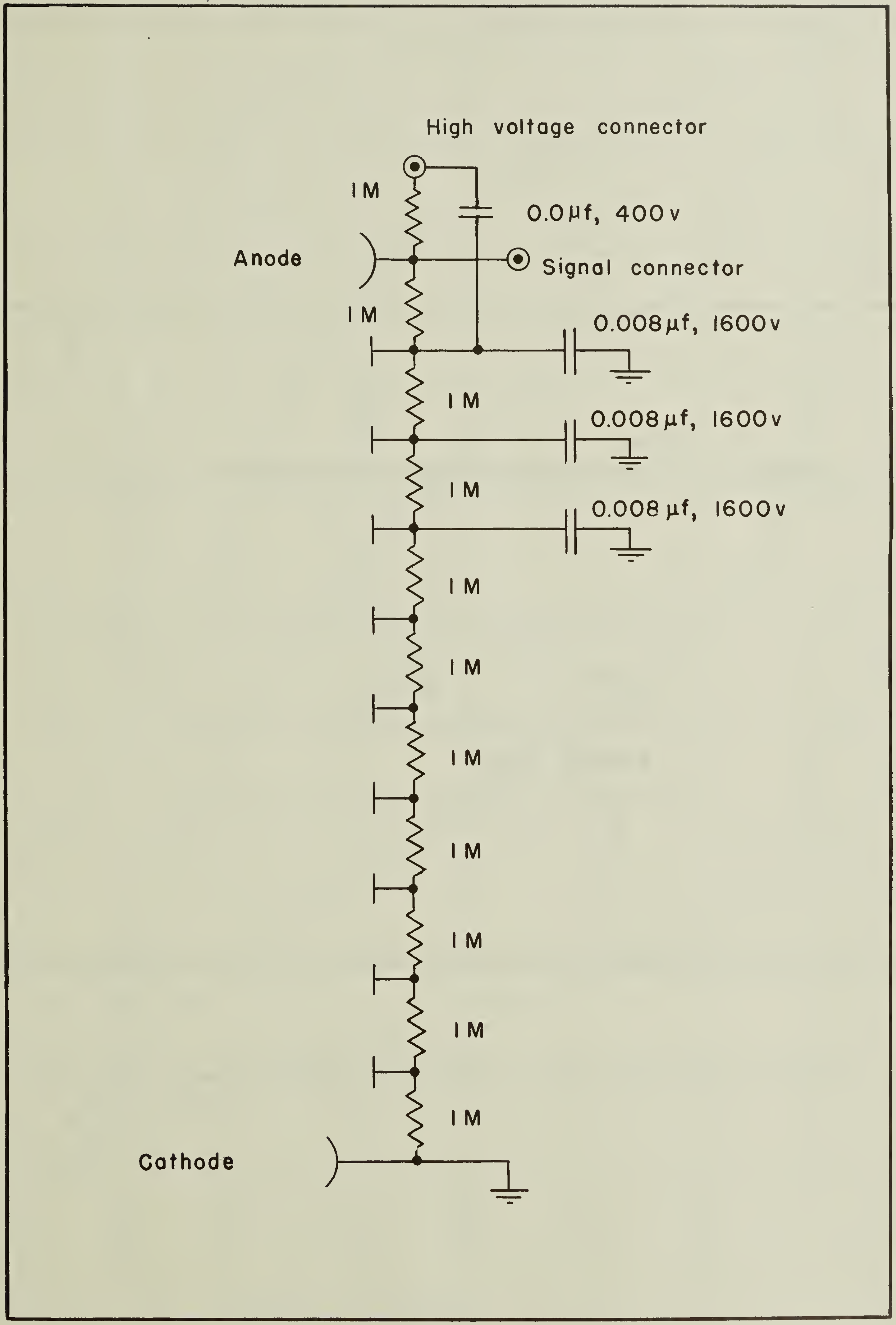

Figure 4. Voltage divider circuit for photomultiplier tube.

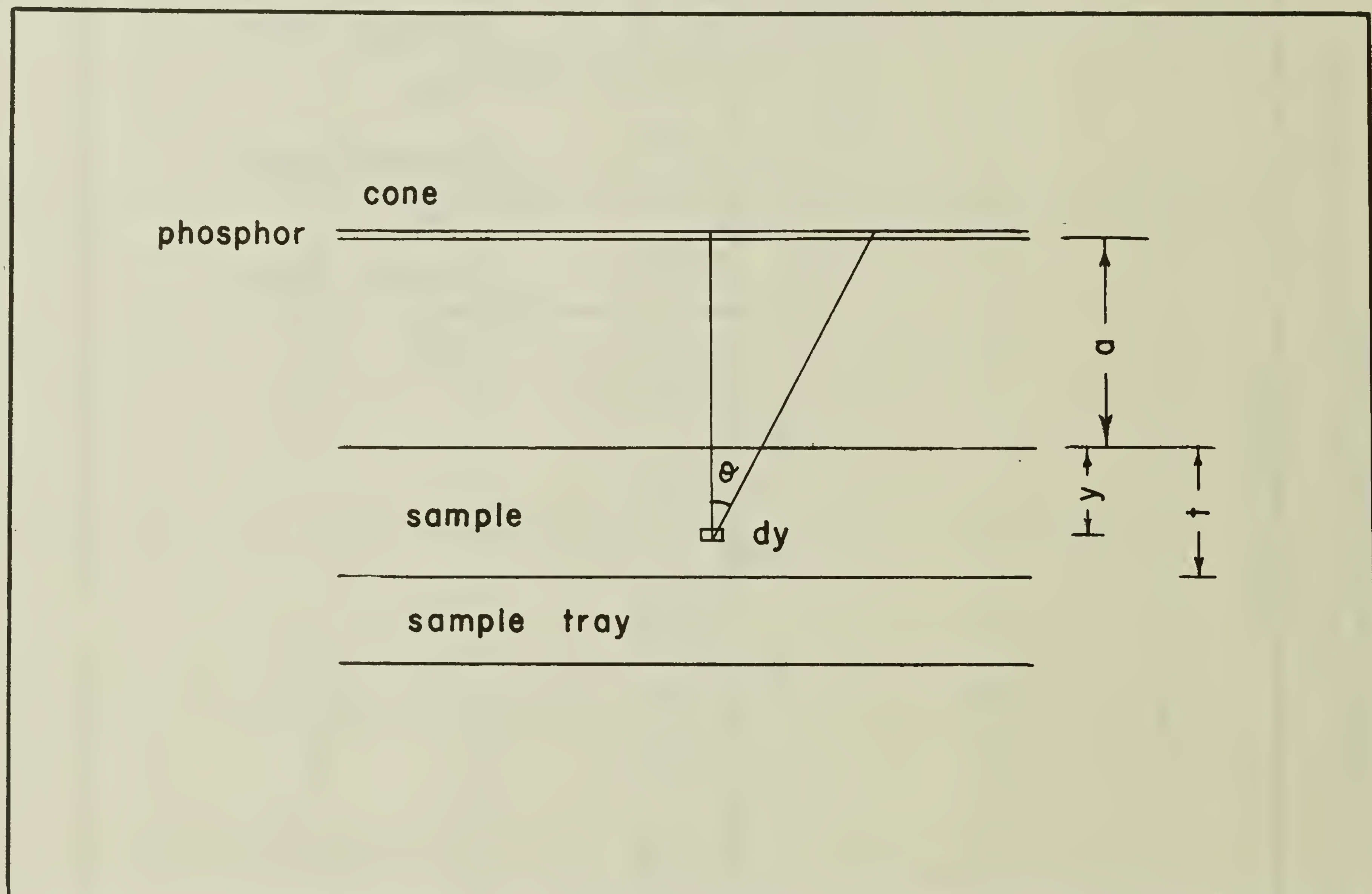

Figure 5. Schematic diagram illustrating relation between a, y, and θ.

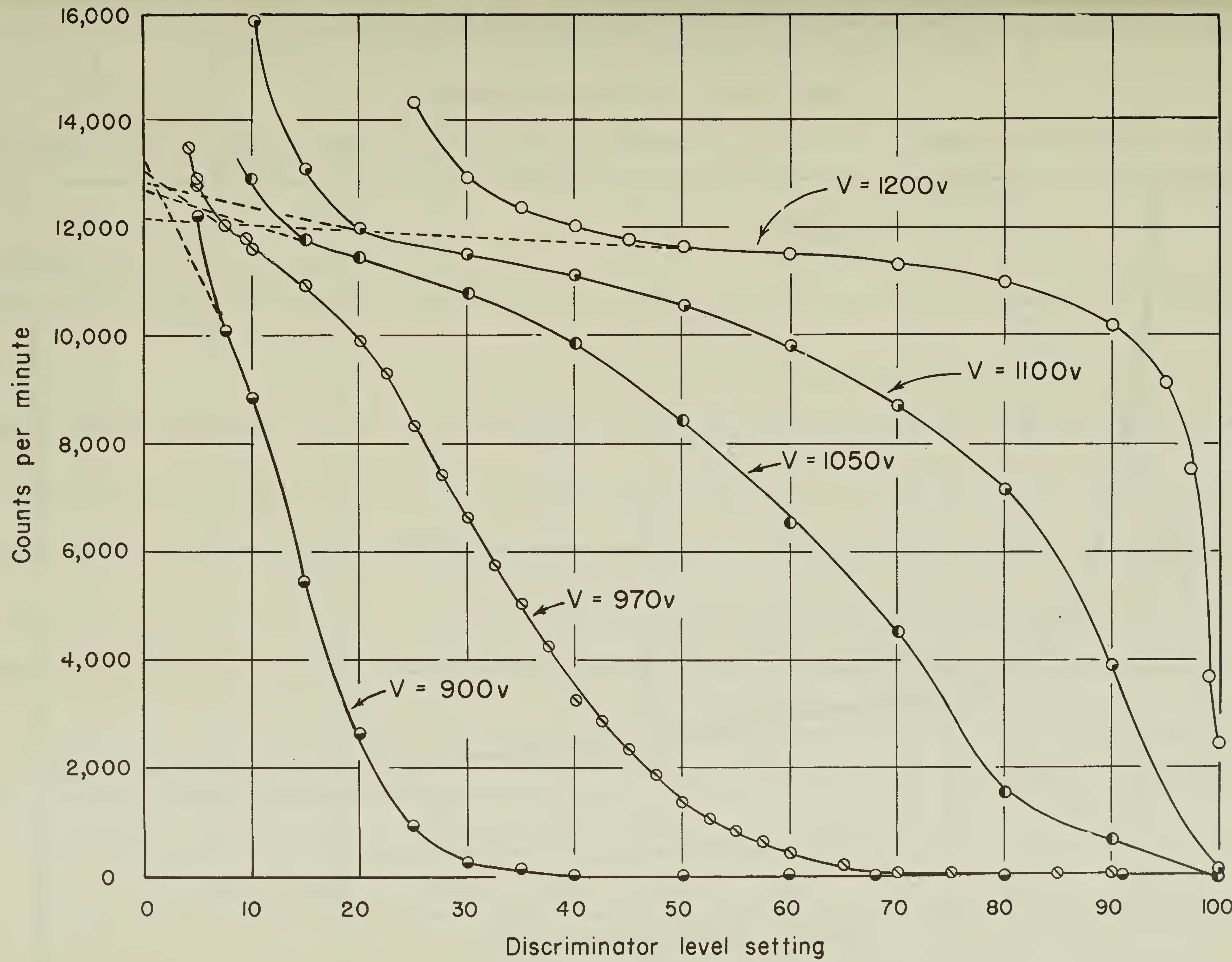

Figure 6. Count rate of a thin polonium source (P-1) as a function of the discriminator level setting at various photo-multiplier assembly voltages.

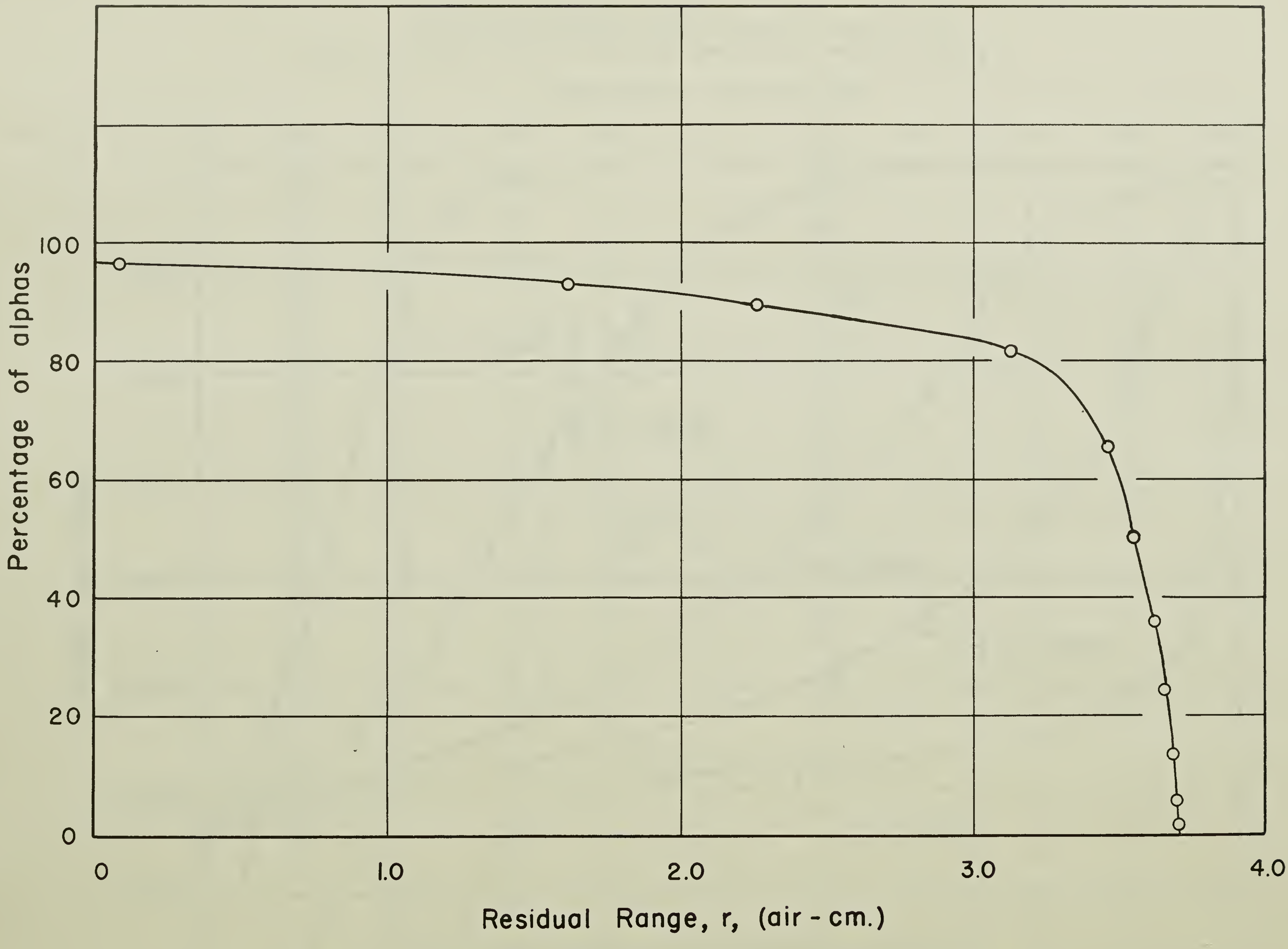

Figure 7. Theoretical distribution of residual alpha particle ranges from a thin source emitting monoenergetic alpha radiation.

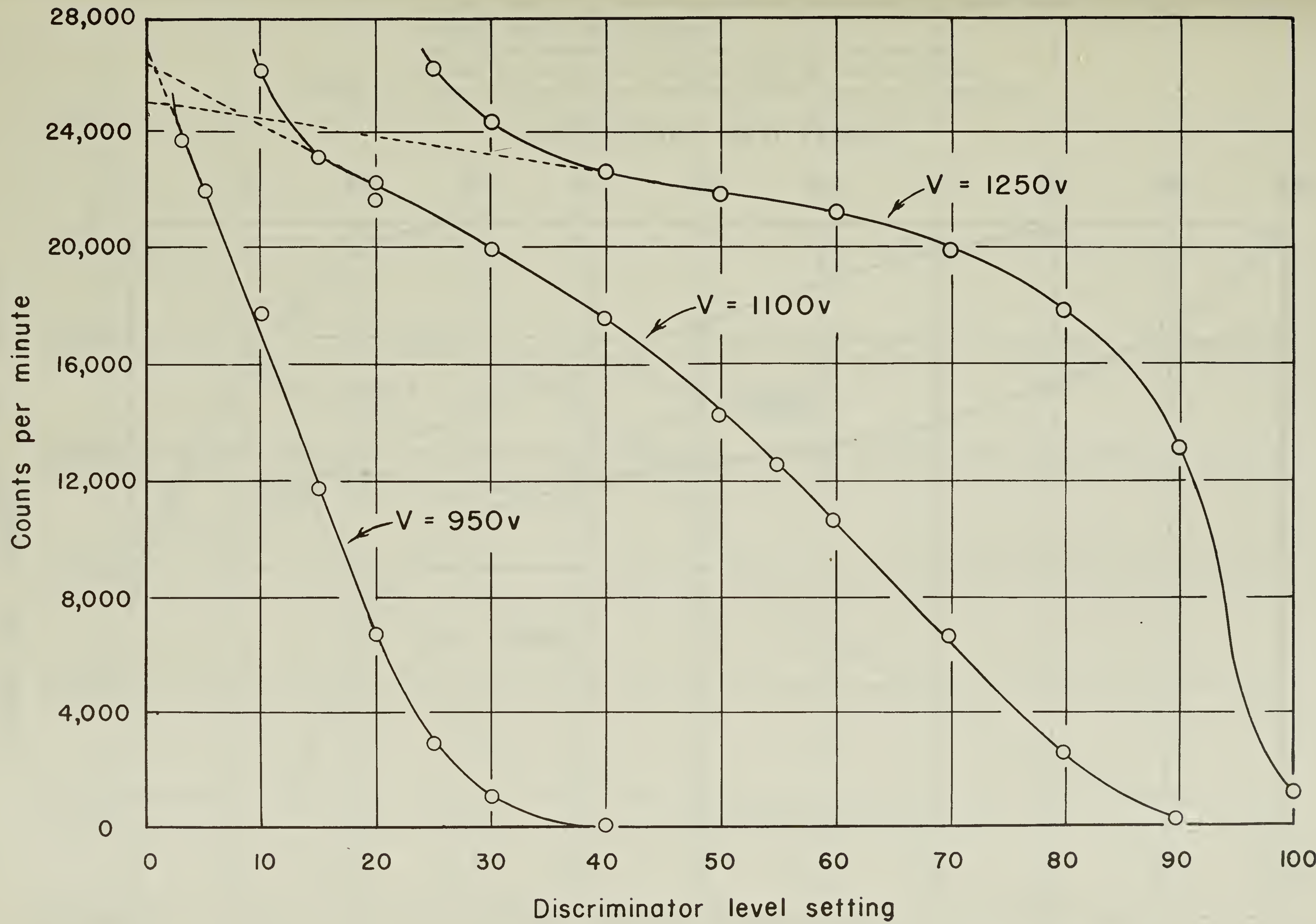

Figure 8. Count rate of a thin polonium source (P-4) as a function of the discriminator level setting at various photomultiplier assembly voltages; no absorber between sample and phosphor.

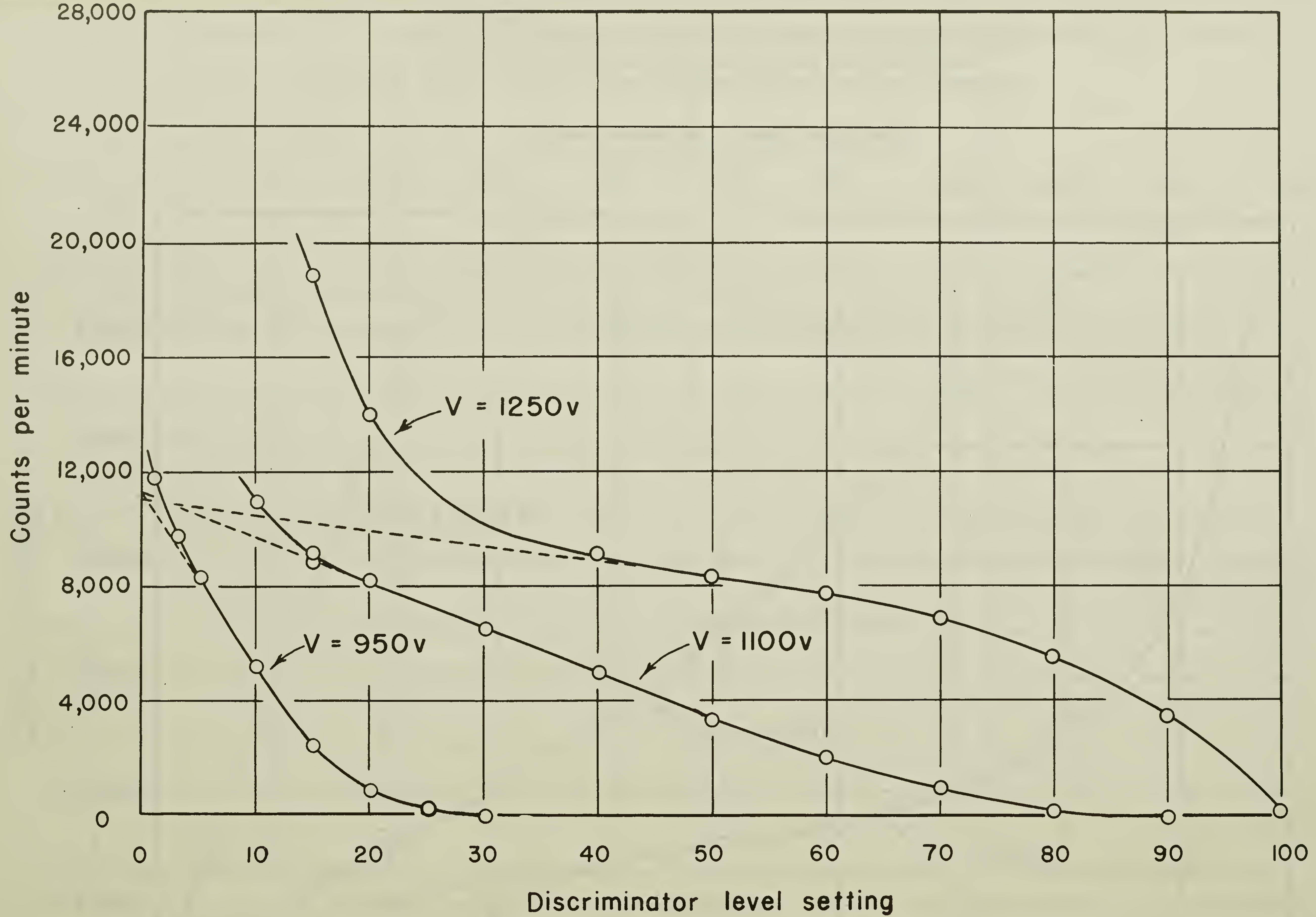

Figure 9. Count rate of a thin polonium source (P-4) as a function of the discriminator level setting at various photomultiplier assembly voltages; 2.45 mg aluminum/cm^2 between sample and phosphor.

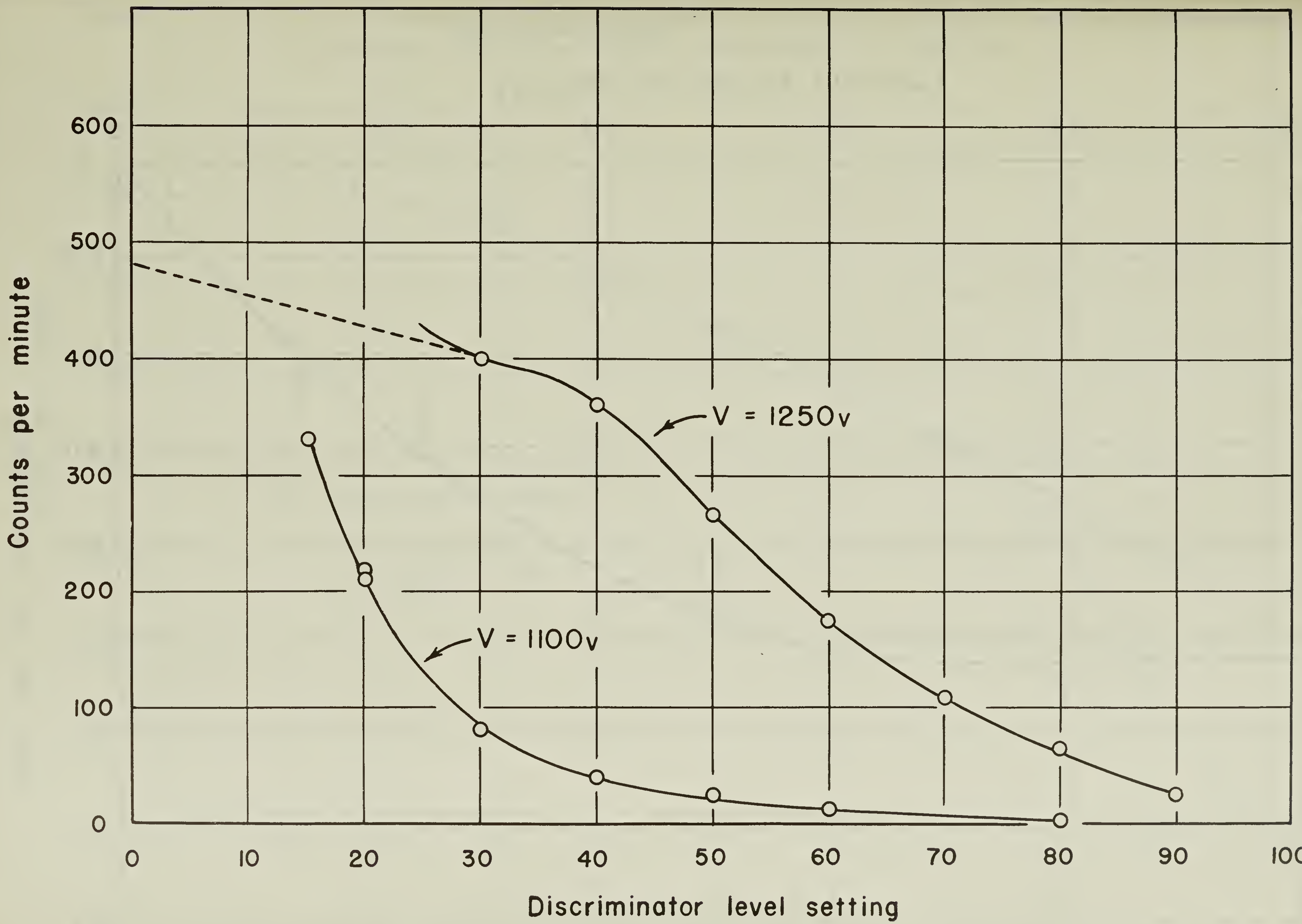

Figure 10. Count rate of a thin polonium source (P-4) as a function of the discriminator level setting at various photomultiplier assembly voltages; 4.90 mg aluminum/cm^2 between sample and phosphor.

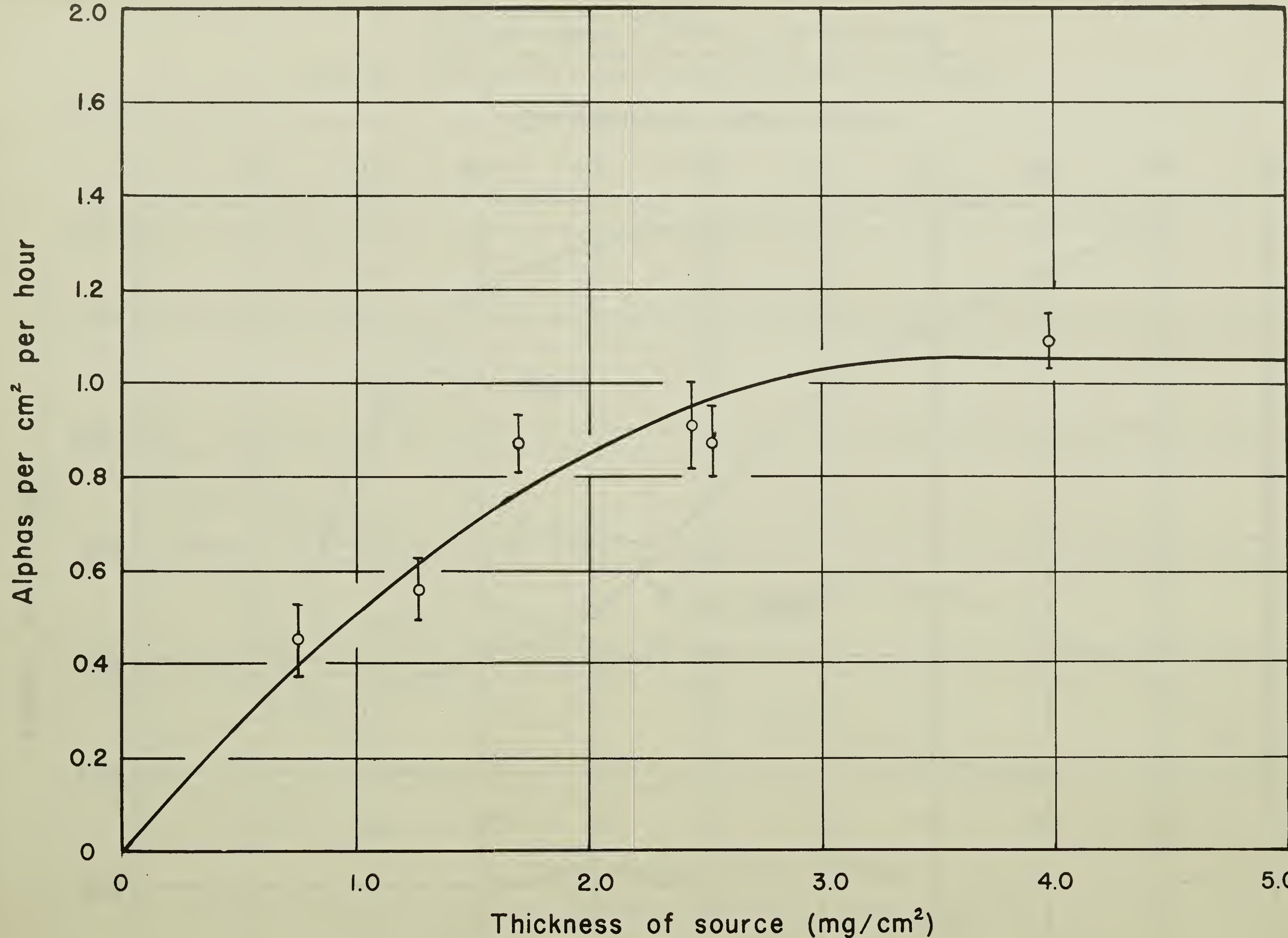

Figure 11. Count rate as a function of thickness of a sample of ocean bottom sediment.

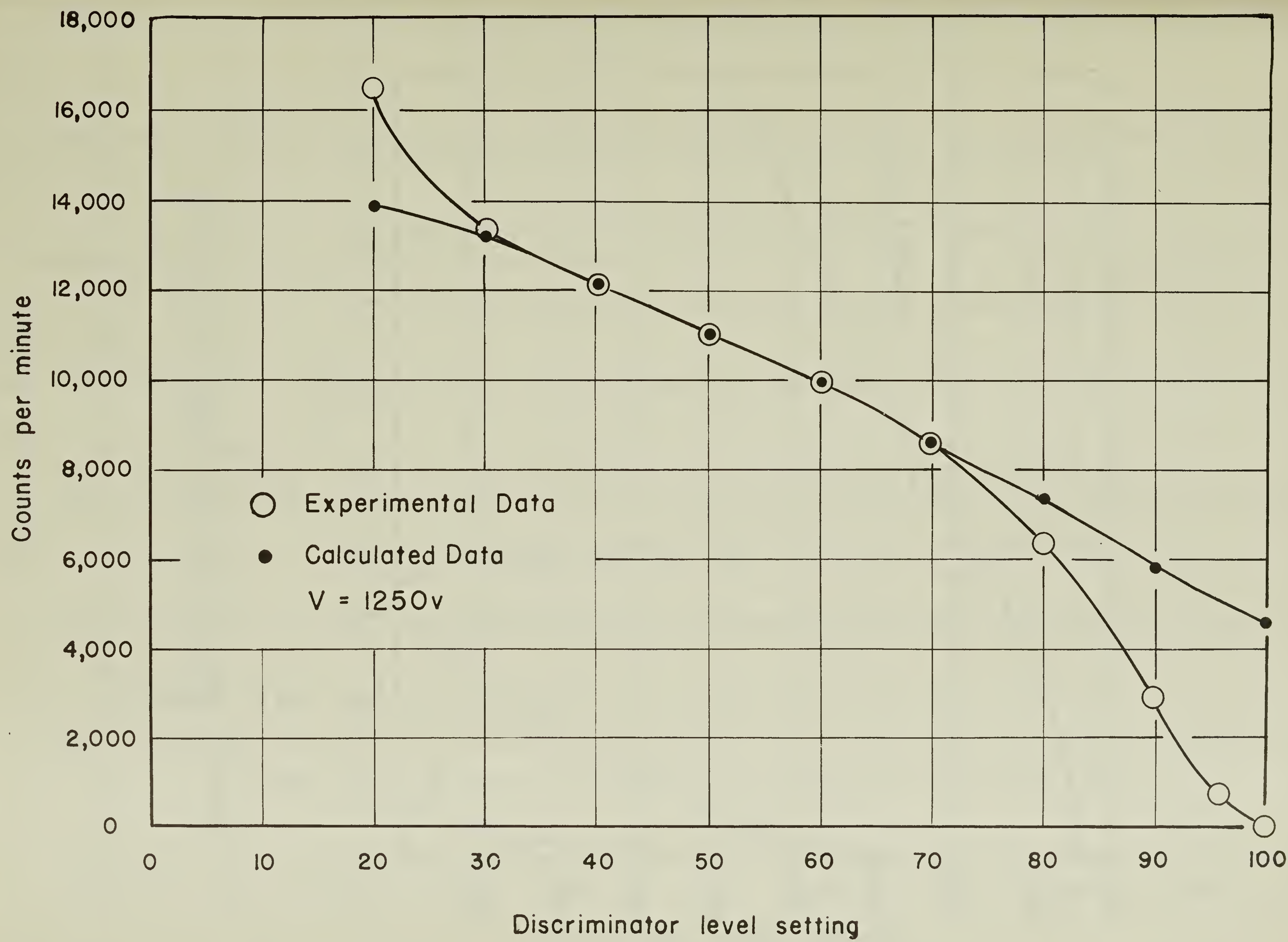

Figure 12. Count rate as a function of discriminator level setting for a thick source of uranium metal strip at a voltage of 1250 volts across the photomultiplier assembly.

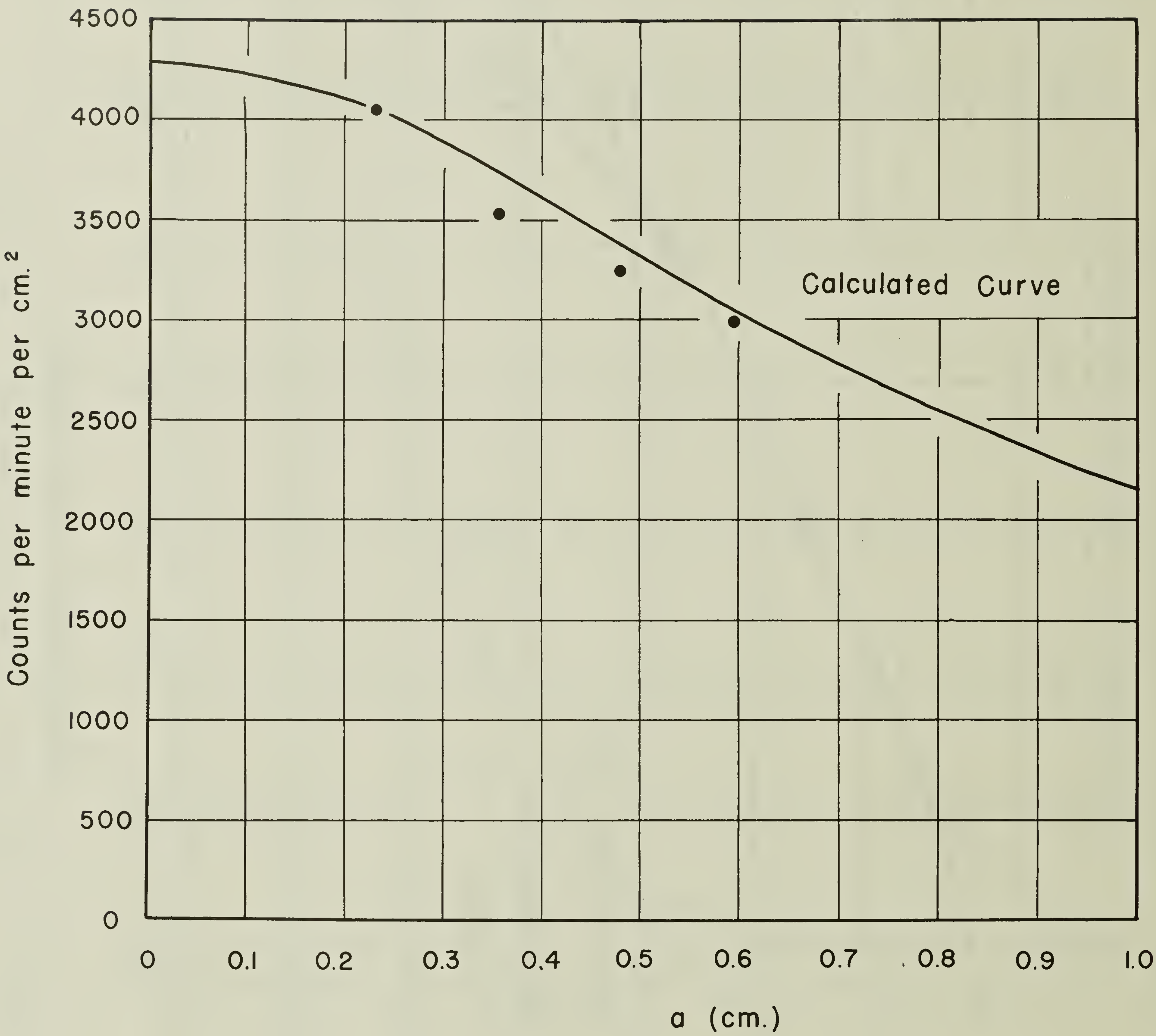

Figure 13. Theoretical and observed count rates for a thick source of uranium metal square as a function of a.

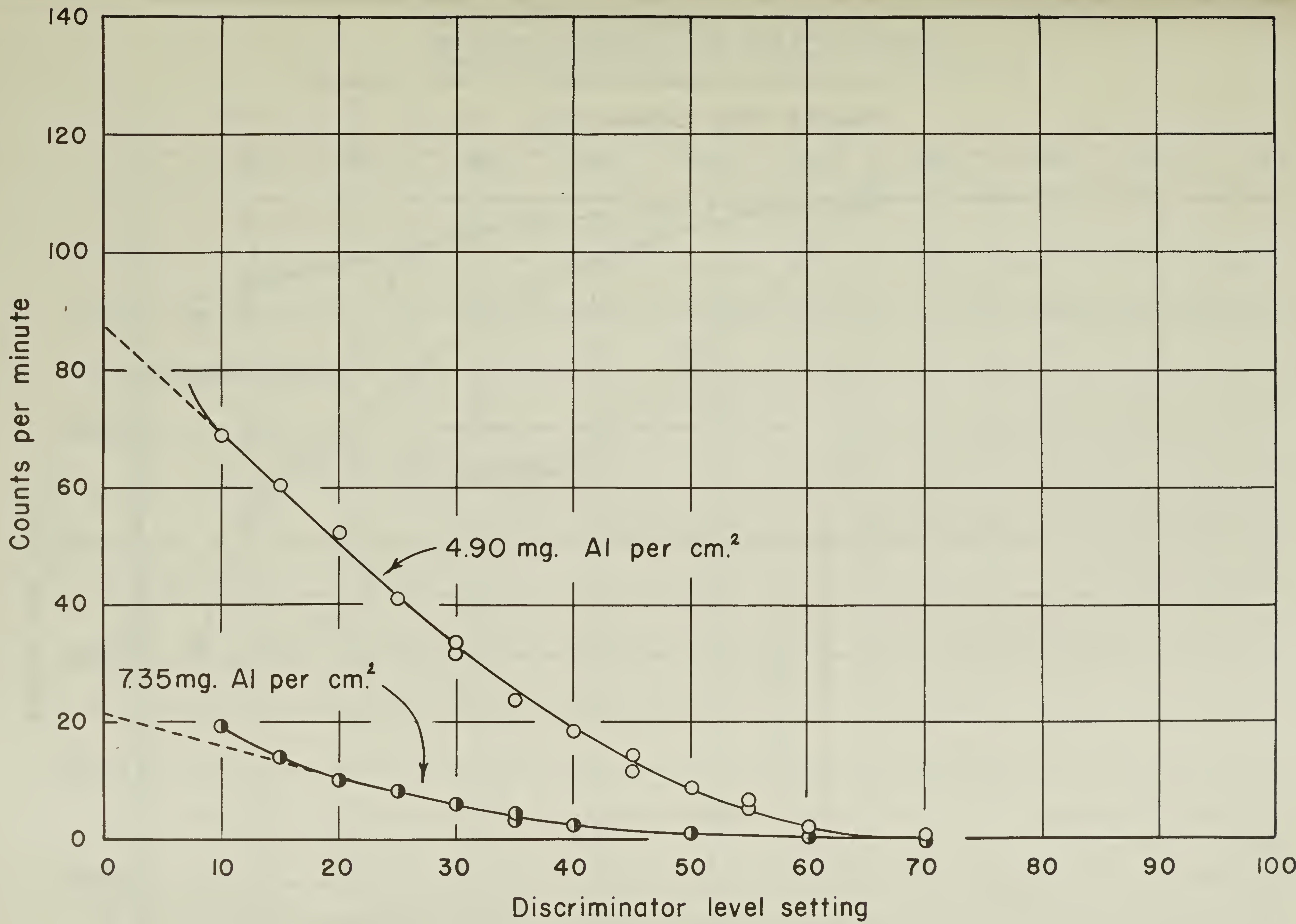

Figure 14. Count rate of a thick source of uraninite (U-d) as a function of discriminator level setting and thickness of absorber at various photomultiplier assemble voltages.

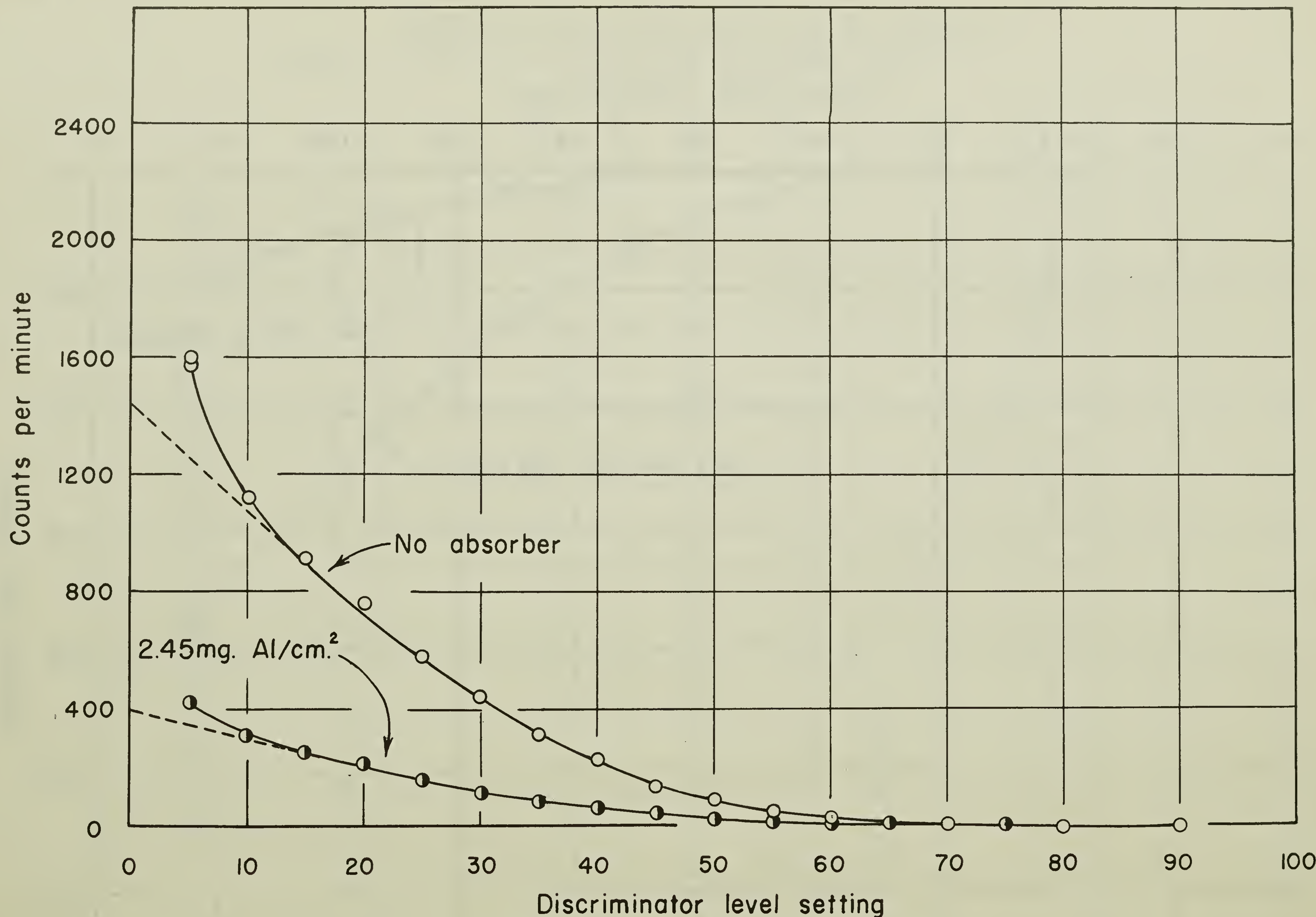

Figure 15. Count rate of a thick source of uraninite (U-d) as a function of discriminator level setting at various photomultiplier assembly voltages; 4.90 mg aluminum/cm^2 between sample and phosphor.

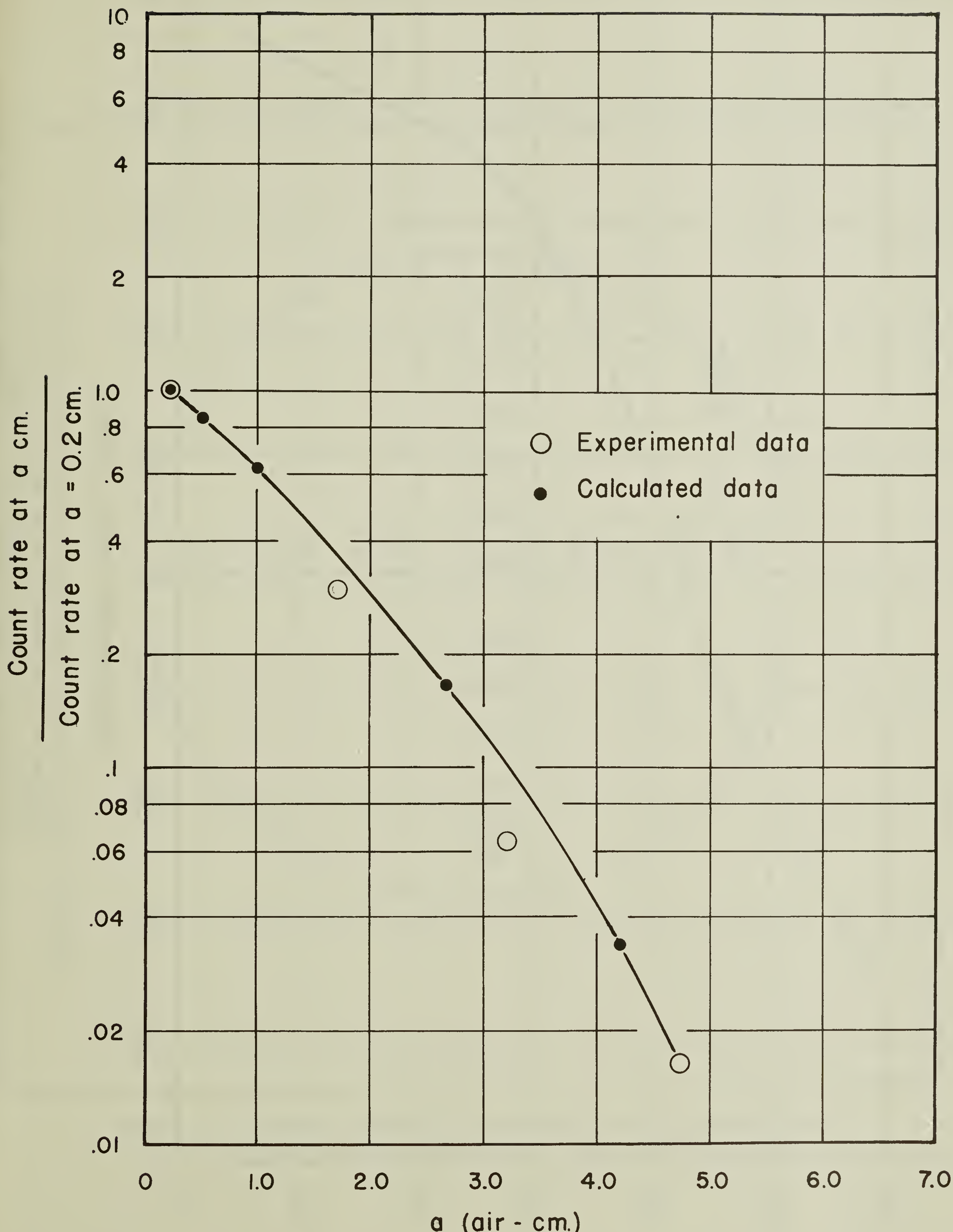

Figure 16. Theoretical and observed count rate of a thick source of uraninite (U-d) at zero discriminator level setting as a function of absorber thickness.

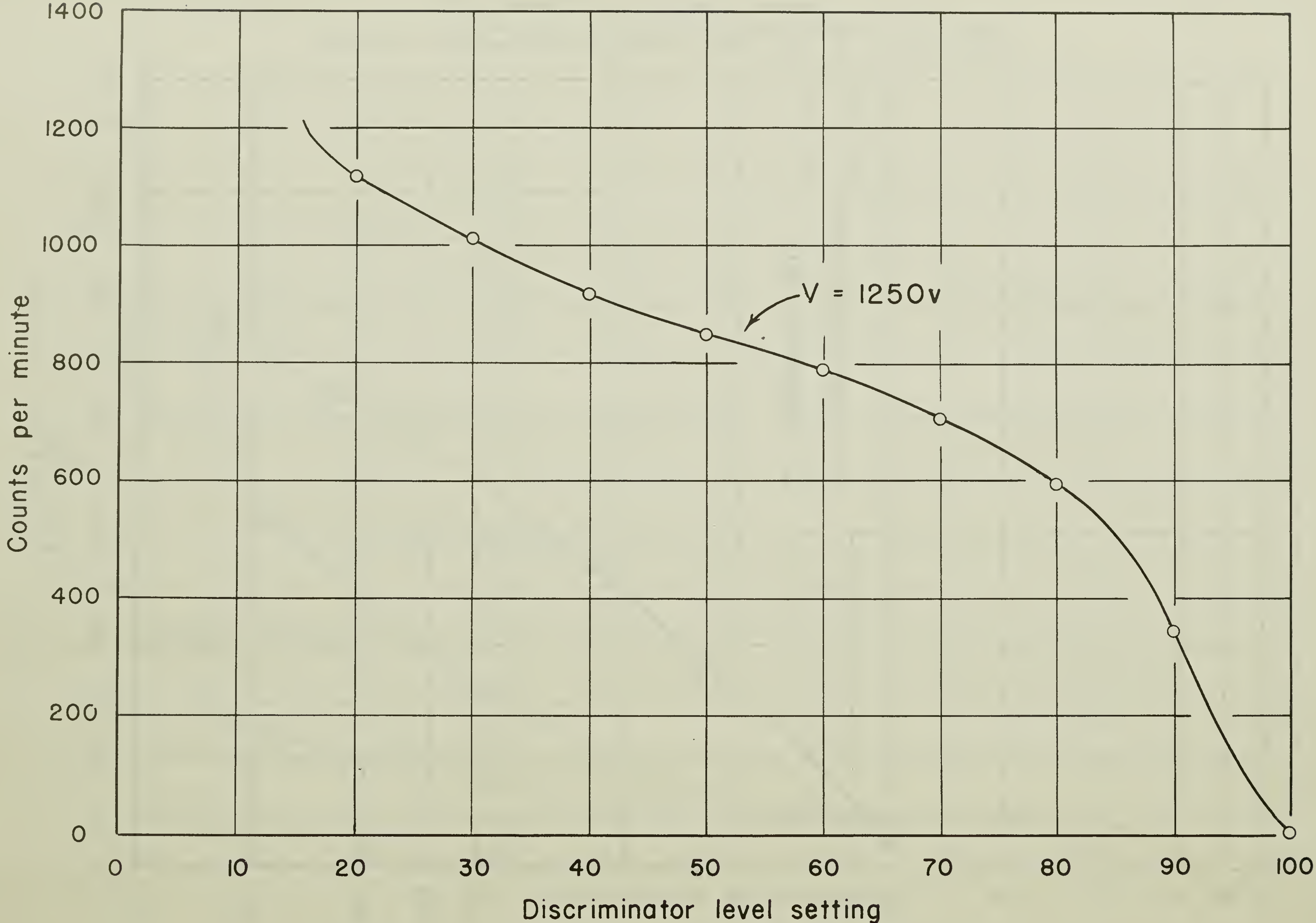

Figure 17. Count rate of a thick source of uraninite (U-d) as a function of discriminator level setting at a voltage of 1250 v. across the photomultiplier assembly.

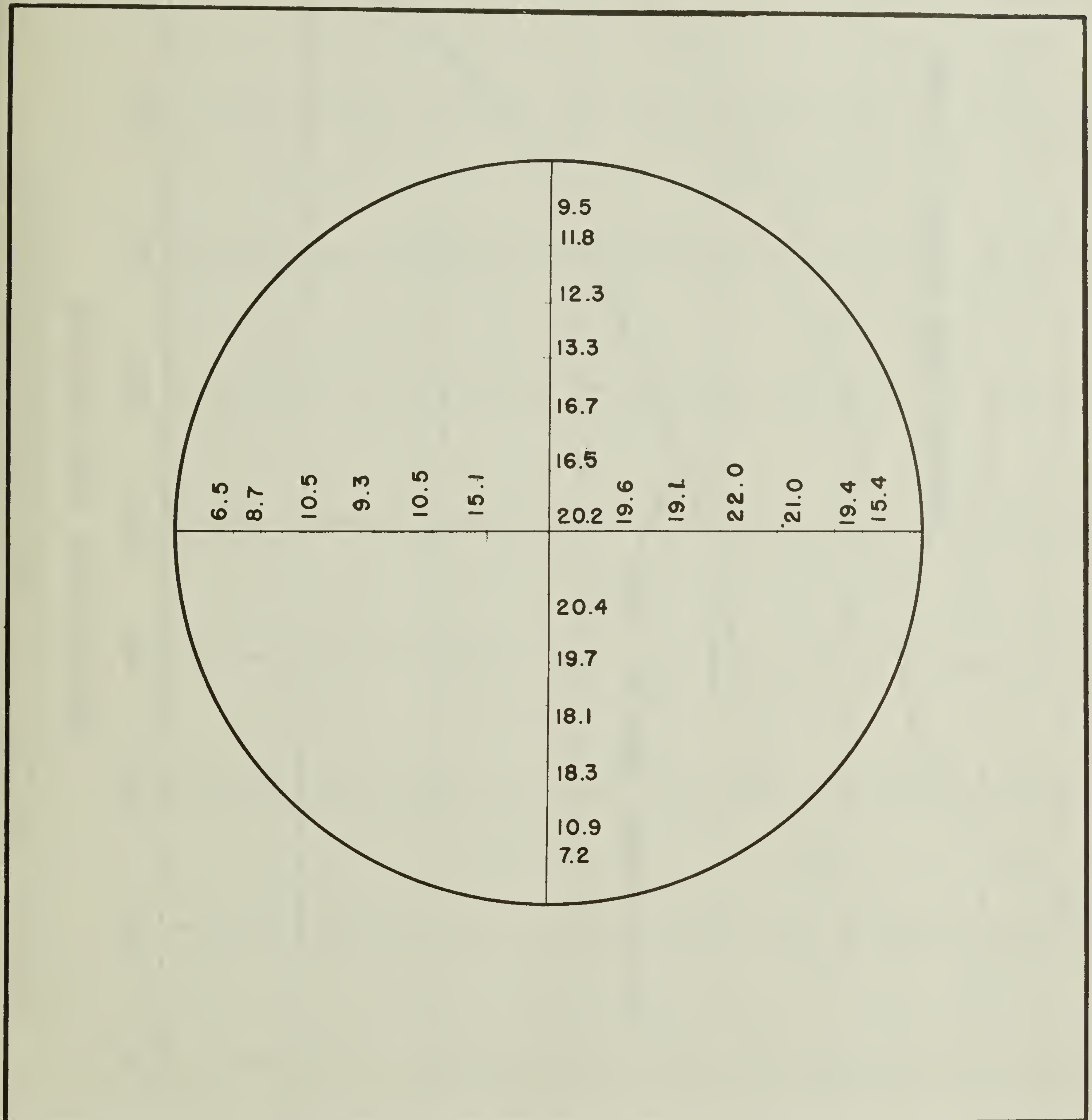

Figure 18. Counting efficiency beneath the cone for a thick source square of uranium metal at a normal operating point.

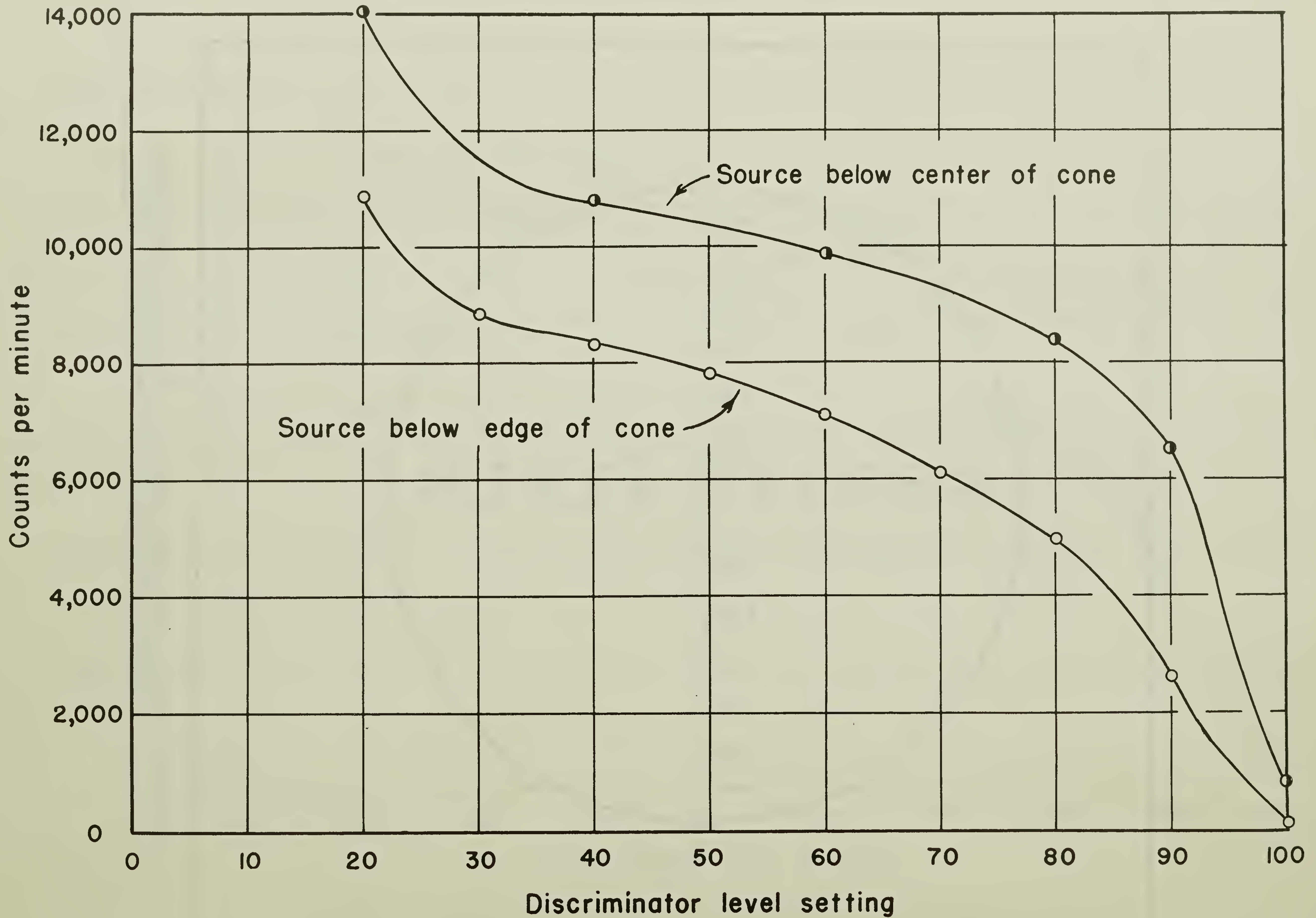

Figure 19. Count rate of a thin source of polonium as a function of discriminator level setting beneath the center and under the edge of the cone.